

REMARKS

A Substitute Specification has been filed herewith as requested by the Examiner. In compliance with the requirements of 37 CFR 1.125, a redlined and unredlined copy has each been filed. As will be apparent from the redlined version of the substitute specification, the specification has been amended to update the status of the parent application at page 1, and to correct several obvious errors appearing as follows with reference to the redlined copy of the substitute specification:

page 3, lines 23-24, reference to claims 2 to 4 has been deleted;

obvious grammatical errors have been corrected at page 5, lines 26 and 31, i.e. "an cathode" has been changed to --a cathode-- and "'such transport or injecting layers" has been changed to --such as transport or injecting layers--;

page 8, line 6 to page 11, line 15: the amendment to the

specification made in the Amendment dated November 1, 2005 has been entered and obvious errors and repetitions inadvertently introduced in said amendment have also been deleted; reference to claims 11 and 12 has been deleted; page 11, line 9, "dimethylhexyloxy" has been changed to -dimethyloctyloxy-- as was done in amendments subsequent to the November 1, 2005 amendment;

page 13, line 25, "from" has been changed to -form-- to correct an obvious typographical error;

page 14, line 9, page 16, line 10, and page 17, line 6, "which" has been inserted for idiomatic readability;

page 16, lines 4 and 17, "not in accordance with the subject matter of the claims" has been restored to the original language --not in accordance with the invention--; and

page 19, line 3, --to-- has been inserted after "well".

The Substitute Specification includes no new matter.

The claims have been amended to more clearly define the invention and to obviate the basis for objection and rejection. Reconsideration of the claims as amended is respectfully requested.

Claims 13 to 24 are now in this case.

The objections and requirements set forth in Paragraphs 5, 6, 7, 8, and 9 of the Office Action dated October 18, 2006 are now moot in view of the present Amendment to the specification and claims.

Claims 17-20 stand rejected under 35 U.S.C. 102 (b) as being anticipated by WO 99/24526, referring to US2002/0064680 as representing a translation of the international application published as WO 99/24526 to Spreitzer et al. The claims as amended are now broader than the claims under consideration in the last Office Action and it is expected that the Examiner will maintain the rejection as anticipated by WO 99/24526. It is submitted that such a rejection is untenable, should be withdrawn for all of the

reasons of record, and all the claims allowed.

Spreitzer et al fail to anticipate the instant claims because, among other reasons, it fails to teach each and every element set forth in the claims. The cited reference is assigned to Aventis Research and grew out of a joint development research agreement between Aventis Research and applicant's Assignee, Koninklijke Philips Electronics. Although there is an overlap of similar subject matter between the publication and the instant application, the disclosures are not identical, the reference fails to teach the exact claimed polymers, the unexpected properties exhibited by the claimed polymers, and the unexpected properties exhibited by EL devices derived from these polymers. The reference includes myriad polymers designated primarily copolymers. Polymers having structures very similar to each other may be grouped as:

Polymers P1, P4, P5, P7, P11, P13, P15, P17, V1, V3, V4, V5, V6, and V7;

Polymers P2, P3, P10, P16, and V2;

Polymers P6 and P8;

Polymer P9;

Polymer P12; and

Polymer P14.

At page 29, the polymers of the reference are said to be prone to the occurrence of unwanted triple and single bonds (or a Tolan-Bisbenzyl defect (TBB) or defective bonds) which adversely affect service life. The TBB of the polymers illustrated in this reference is given in Table 2. It should be noted that Polymer V2 (included among the polymers disclosed to be comparative examples not in accordance with the invention of Spreitzer et al) has a description that is suggestive of both polymers claimed herein and other polymers P2, P3, P10, and P16 disclosed in Spreitzer et al; however, Polymer V2 has a TBB and service life that is undesirable

according to Spreitzer et al. On the other hand, Polymers V5 and P4 have a description structure that is suggestive of polymers that are not in accordance with the present invention and are not comparable to the polymers of this invention in enhancing the service life of an EL device (see comparative example 1 of the present specification, for example) but according to the reference have a TBB and service life that are highly desirable therein. Similarly, Polymers P6 and P8 having a description that is suggestive of polymers claimed herein barely make Spreitzer et al.'s the patentee's range of satisfactory TBB, and Polymers P9 and P12 having a description that is suggestive of polymers claimed herein are in Spreitzer et al.'s undesirable range of TBB, indicating relatively poor service life properties according to Spreitzer et al. Thus the reference appears to teach polymers that exhibit properties /results that are diametrically opposed to the properties/results obtained and claimed herein.

It has long been recognized that structural formulae and descriptions of polymers are imprecise and cover a vast number of products. It has also long been held that a chemical compound is not limited to a formula or name but must be considered in the light of all its properties. See *In re Papesch*, 315 F.2d 381, 391, 137 USPQ 43, 51 (CCPA 1963) ("From the standpoint of patent law, a compound and all its properties are inseparable."). ("The fact that a claimed compound may be encompassed by a disclosed generic formula does not by itself render that compound obvious."); *In re Jones*, 958 F.2d 347, 350, 21 USPQ2d 1941, 1943 (Fed. Cir. 1992) (Federal Circuit has "decline[d] to extract from *Merck [& Co. v. Biocraft Laboratories Inc.*, 874 F.2d 804, 10 USPQ2d 1843 (Fed. Cir. 1989)] the rule that... regardless of how broad, a disclosure of a chemical genus renders obvious any species that happens to fall

within it."). See also *In re Deuel*, 51 F.3d 1552, 1559, 34 USPQ2d 1210, 1215 (Fed. Cir. 1995). Moreover, a *prima facie* case of obviousness based on structural similarity is rebuttable by proof that the claimed compounds possess unexpectedly advantageous or superior properties. *In re Papesch*, 315 F.2d 381, 137 USPQ 43 (CCPA 1963).

Further, it is well established that a claim is anticipated only if each and every element as set forth in the claim is found, either expressly or inherently described, in a single prior art reference." *Manual of Patent Examining Procedure* § 2131 (8th ed., Rev. 4, Oct. 2005), citing *Verdegaal Bros. v. Union Oil Co. of California*, 814 F.2d 628, 631, 2 U.S.P.Q. 2d 1051, 1053 (Fed. Cir. 1987). The standard for rejection under 35 U.S.C. § 102 is identity. WO 99/24526 in particular does not disclose the service life limitations now recited in all of the claims, e.g., that the organic electroluminescent device is capable of providing a service life of at least 45h when driven at a constant current, at an initial brightness of 200 Cd/m², and at an ambient temperature of 80 °C. (claims 17-27) or the electroluminescent device is capable of providing a service life of at least 800h when driven at a constant current, at an initial brightness of 100 Cd/m², and at an ambient temperature of 70 °C (claim 29). The reference fails as an anticipatory reference.

The Examiner deems the service life limitation of the instant claims to be inherent in the prior art devices made with polymer 9 (and we assume for the sake of argument that the same would be asserted as to polymers P6, P8, and P12). Yet as pointed out above, based on patentee's description of the TBB content and its effect on service life, the contraindications of the reference when compared to the instant invention and vice versa, it cannot be said with certainty that the claimed service life limitation is inherent in the prior art polymers.

Inherency arises when a single prior art reference fails to disclose the claimed invention per se, but the natural and invariable practice of the reference would necessarily and inherently meet all of the elements of the claimed invention. *Ethyl Molded Products v. Betts Package, Inc.*, 9 U.S.P.Q. 2d. 1001, 1032-1033. Inherency exists only when the prior inherent event can be established as a certainty; that an event may result from a given set of circumstances is not sufficient to establish anticipation by inherency. See *Phillips Petroleum Co. v. U.S. Steel*, 6 U.S.P.Q.2d 1065 at 1076-1077, 673 F. Supp. 1278 (D. Del. 1987).

Probabilities are not sufficient for prior art to anticipate an invention inherently; a prior inherent event cannot be established based on speculation or where a doubt exists. *Id.*; *E.I. du Pont v. Phillips Petroleum*, 2 U.S.P.Q.2d 1545 at 1552, 849 F. 2d 1430 (Fed. Cir. 1988); *Schering Corp. v. Precision-Cosmel Co.*, 227 U.S.P.Q. 278 614 F. Supp. 1368 (D. Del. 1985) and many other cases.

The instant claims are directed to an organic electroluminescent device which is capable of providing a service life of at least 45 h when driven at a constant current, at an initial brightness of 200 Cd/m², and at an ambient temperature of 80 °C.

None of these highly specific features, which are service life characteristics that were experimentally determined and are required of the device of the instant claims, could have been established as a certainty without any doubt, for the claimed polymers and/or the claimed device, based on what is shown in WO 99/24526. The instant claims are not inherently anticipated because they are directed to subject matter which is different from and is therefore not expressly anticipated by WO 99/24526, and this subject matter, the service features, could not have been predicted, absent improper reference to Applicant's disclosure.

Speculation based on mere structural or descriptive

similarity is insufficient to establish inherent anticipation, in view of a legal standard that requires absence of any doubt.

For the foregoing reasons, the rejection of claims 17-20 as anticipated directly or inherently under 35 U.S.C. §102(b) should be withdrawn, an action that is respectfully requested.

Claims 13 to 20 stand rejected as unpatentable under 35 U.S.C. §103(a) in light of reference WO 99/21936. Reconsideration of the claims as amended is requested.

WO99/21936 is assigned to applicant's Assignee, Koninklijke Philips Electronics, and indeed there is a common inventor between the present application and the PCT publication, both applications being directed to subject matter that grew out of the same research project. Again, although there is an overlap of similar subject matter between the publication and the instant application, the disclosures are not identical and the reference fails to teach the exact polymers, the unexpected properties exhibited by the polymers, and the unexpected properties exhibited by EL devices derived from these specific polymers. The reference relates to electroluminescent (EL) devices comprising a soluble poly(1,4-phenylenevinylene) but it does not disclose or suggest the polymers claimed herein nor is there disclosure or suggestion of the claimed ability of the polymers to enhance the service life of EL devices in which they are incorporated. Although the reference discloses a formula for a repeating unit C1 at page 4, no disclosure of polymers as claimed herein is found. The reference does not disclose any polymers comprising or consisting of units of C1 wherein $p=1, 2$ or 3 and appears to be limited to polymers where $p=0$. See Example 2 and Table 1 on pages 16 and 17 wherein R^4 is H in every instance. It is nevertheless contended that the claims are obvious despite the lack of disclosure.

Objective evidence or secondary considerations such as

unexpected results, commercial success, long-felt need ... are relevant to the issue of obviousness and must be considered in every case in which they are present." [emphasis added] *Manual of Patent Examining Procedure*, §2141, p. 97 (8th Ed. Rev.2, May 2, 2004).

Applicants' specification provides Examples 1 and 2, showing output comparisons of prior art copolymers of WO 99/21936 with polymers of the present claims. See Examples 1 and 2 pp. 9-14 of the specification as originally filed. The specification as originally filed thus subjected an electroluminescent (EL) device of WO 99/21936 to a service life test in which the device, the positive pole of the voltage source being connected to an anode and the negative electrode to a cathode, was driven at constant current and at an initial brightness of 100 Cd/m² while maintaining an ambient temperature of 70 °C. See p. 10 lines 9-12 in the specification as originally filed. The service life, defined as the time within which the brightness drops to half its initial value, thus determined was found to be about 180 h. See p. 10 lines 12-13 in the specification as originally filed. The voltage increase needed to maintain a constant current during the service life was found to be about 0.015 V/h. See p. 10 lines 13-14 in the specification as originally filed. The service life at 20 Cd/m² and room temperature exceeds 5000 h, which demonstrates the drastic reduction of service life associated with the use of a higher brightness and an elevated ambient temperature of 70 °C. See p. 10 lines 14-17 in the specification as originally filed.

In comparison, when the EL devices including the monomers of the present claims (see for example formula C5 and C6) were subjected to the same service life test, the service life of the EL device comprising the polymer of formula (C5) was 925 h. The voltage increase was about 2 mV/h. By collecting the data for a large number of samples the service life is found to be at least 800 to 1200 h. See p. 11 lines 12-15 of the specification as originally filed. Similarly, the service life of the EL device

comprising the polymer of formula (C6) was found to be at least 800 to 1200 h. The voltage increase was found to be about 1.5 to 2 mV/h. See p. 11 lines- 16-17 of the specification as originally filed. See other comparative examples in the specification as well. All of the instant claims now recite the service life enhancing properties of the polymers or the service life properties of the EL device as a limitation.

The Examiner appears to take the position that that the service life limitations are unpersuasive because Claim 17 does not limit the voltage at which the initial brightness value is obtained stating that it is well known in the art and admitted by applicants on page 1 of the present application that brightness can be increased by increasing voltage. However, this rationale totally omits the fact that it is also well known that increasing the voltage also decreases the service life. In discussing WO 99/21936, the present application at page 1 asserts:

"Aryl-substituted poly-p-arylenevinylenes of the type mentioned in the opening paragraph and electroluminescent (EL) devices, polymer electroluminescent devices for short, comprising such polymers are disclosed in the international patent application WO 99/21936. Dependent on the particular structure, the polymers disclosed therein are capable of light emission in the green to red range of the visible spectrum. The color of light emission does not shift when a polymer EL device comprising the known polymer is stored at elevated ambient temperatures for an extended period of time. Furthermore, the polymer EL devices comprising these polymers have a service life exceeding 1500 to 5000 h when driven at a brightness of 20 Cd/m². For many purposes however, a brightness of 20 Cd/m² is too low and a brightness of at least 100 to 200 Cd/m² is required. This is for example the case in a matrix display device which is driven in a time-multiplexed manner. Although the brightness of an EL device can be simply increased by increasing the voltage at which it is driven increasing the brightness generally leads, as is well-known in the art, to a substantial reduction in service life. A further substantial reduction in service life is generally observed when a polymer EL device is driven at elevated ambient temperatures such as 70 to 80 °C. However, satisfactory

operation of the devices at those elevated temperatures for an extended period of time is a prerequisite for many applications. Such applications include display devices for automotive applications and mobile phones." (emphasis added).

It is applicant's discovery that certain aryl-substituted poly-p-arylenevinylenes as claimed herein have the ability to enhance the service life properties of EL devices comprising them and that said enhancement is not reduced by increasing the brightness or when the EL device is driven at a constant current, at an initial brightness of 200Cd/m², and at ambient temperatures such as 70 to 80 °C. The rejection is untenable and should be withdrawn.

In view of the above, it is respectfully submitted that the present application is in condition for allowance, and a Notice of Allowance is earnestly solicited.

Respectfully submitted,

By/Ernestine C. Bartlett/
Ernestine C. Bartlett Reg.No.22,861
Attorney for Applicant(s)

Aryl-substituted poly-p-arylenevinylenes

This application is a continuation of Application Serial No. 09/790,201, filed February 21, 2001, now U.S. Patent No. 6,743,525.

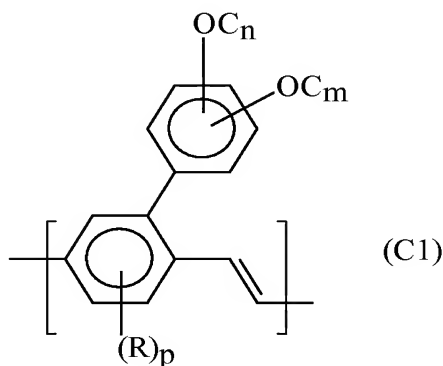
The invention relates to aryl-substituted poly-p-arylenevinylenes, the use thereof in organic electroluminescent devices and organic electroluminescent devices comprising such aryl-substituted poly-p-arylenevinylenes.

Aryl-substituted poly-p-arylenevinylenes of the type mentioned in the opening paragraph and electroluminescent (EL) devices, polymer electroluminescent devices for short, comprising such polymers are disclosed in the international patent application WO 99/21936. Dependent on the particular structure, the polymers disclosed therein are capable of light emission in the green to red range of the visible spectrum. The color of light emission does not shift when a polymer EL device comprising the known polymer is stored at elevated ambient temperatures for an extended period of time. Furthermore, the polymer EL devices comprising these polymers have a service life exceeding 1500 to 5000 h when driven at a brightness of 20 Cd/m². For many purposes however, a brightness of 20 Cd/m² is too low and a brightness of at least 100 to 200 Cd/m² is required. This is for example the case in a matrix display device which is driven in a time-multiplexed manner. Although the brightness of an EL device can be simply increased by increasing the voltage at which it is driven increasing the brightness generally leads, as is well-known in the art, to a substantial reduction in service life. A further substantial reduction in service life is generally observed when a polymer EL device is driven at elevated ambient temperatures such as 70 to 80 °C. However, satisfactory operation of the devices at those elevated temperatures for an extended period of time is a prerequisite for many applications. Such applications include display devices for automotive applications and mobile phones.

It is an object of the invention to provide, inter alia, aryl-substituted poly-p-arylenevinylenes which, when used in an organic EL device, render the EL device capable of providing a satisfactory service life when driven at an initial brightness of at least 100 or better still, at least 200 Cd/m². More in particular the EL device should be capable of

providing a satisfactory service life even in the case where the EL device is driven at elevated ambient temperatures of 70 to 80 °C.

In accordance with the invention, this objective is achieved by aryl-substituted poly-p-arylenevinylenes comprising a repeating unit of the formula (C1),



- 5 in which one or more of the unsubstituted aromatic carbon atoms may be replaced by nitrogen atoms, $-OC_m$ and $-OC_n$ are alkoxy groups, m and n are integers from 2 to 6 with $m + n = 8$, p is 0, 1, 2 or 3 and in which R is CN, Cl, F, CF_3 , NO_2 , or SO_3Z wherein Z is a monovalent cation such as Na^+ , or in which R is $-XR^1$ wherein the unit $-X-$ represents a single bond, $-O-$, $-S-$, $-CO-$, $-COO-$, $-OCO-$, $-SO-$, $-SO_2-$, $-N(R^2)-$ or $-N(R^2)CO-$, and wherein
- 10 R^1 and R^2 are the same or different and constitute a straight-chain branched or cyclic C_1-C_{20} alkyl group or together an C_1-C_{20} alkylene group, in which C_1-C_{20} alkyl or C_1-C_{20} alkylene group one or more hydrogens are optionally substituted by F or a C_4-C_{12} aryl group and/or one or more non-adjacent $-CH_2-$ units are optionally substituted by C_4-C_{12} arylene, $-O-$, $-S-$, $-CO-$, $-COO-$, $-OCO-$, $-SO-$, $-SO_2-$, $-N(R^3)-$ or $-N(R^3)CO-$ where R^3 is C_1-C_{20} alkyl, or in
- 15 which R is a C_4-C_{12} aryl group which may or may not be substituted.

As a typical example, an organic EL device comprising a yellow to green light emitting polymer in accordance with the invention as electroluminescent material is capable of providing a service life of at least 45 to 200 h when subjected to an accelerated service life test which involves driving the device at a constant current, at an initial brightness of 200

20 Cd/m^2 and at an ambient temperature of 80 °C.

An EL device comprising a red to orange light emitting polymer in accordance with the invention as electroluminescent material in an organic EL device is at least capable of providing a service life of at least 800 to 1200 h when subjected to an accelerated service life test which involves driving the device at a constant current, at an initial brightness of 100

25 Cd/m^2 and at an ambient temperature of 70 °C.

In the context of the invention, the service life is defined as the time at which the brightness is reduced to half its initial value while driving the device at a constant current. Evidently, such a device is also capable of operating that long at a constant brightness of at least half said initial brightness.

5 In the context of the invention, the term organic includes polymeric whereas the term polymer and affixes derived therefrom, includes homopolymer, copolymer, terpolymer and higher homologues as well as oligomer.

In WO 98/271376, EL polymers similar to the polymers of WO 99/21936 are disclosed.

10 Preferably, the polymers in accordance with the invention are used as electroluminescent polymers. Alternatively, they may be suitably used as photoluminescent, charge-injecting and/or charge-transporting polymers. In case of an EL device comprising a single organic layer comprising the polymer in accordance with the invention, the polymer serves as charge-injecting charge-transport and electroluminescent polymer at the same time.

15 Referring to formula (C1), p is preferably 0 or 1 and R, if present, is located at a 5-position of the phenylene ring.

If the polymer is to emit green or more greenish light p is preferably 0 and R, if present, selected to be a C₁-C₂₀ alkyl group of the type defined above.

20 If the polymer is to emit yellow or more yellowish light the R is preferably selected to be a C₁-C₂₀ alkoxy group of the type defined above. Preferred alkoxy groups are OC₄, in particular 2-methylpropyloxy, and OC₈, in particular 3,7-dimethylhexyloxy. Even more preferred is methoxy.

~~Preferred embodiments of the polymers in accordance with the invention are claimed in claims 2 to 4.~~

25 In another preferred embodiment, the polymer is characterized in that the proportion of repeating units which are repeating units of formula (C1) is at least 0.95. The benefit in terms of service life to be had by using the polymers in accordance with the invention is optimized if the proportion of repeating units of formula (C1) is as large as possible. Preferred is a proportion of at least 0.95 because then the emission spectrum of the copolymer or higher homologue substantially coincides with that of the homopolymer of
30 repeating units according to formula (C1).

In yet another preferred embodiment, the polymer is characterized in that the proportion of repeating units which are repeating units of formula C1 is less than 0.95. In order to widen the range of properties available within the class of polymers in accordance

with the invention, which may for example be advantageous if the emission spectrum of the polymer is to be adapted to suit the needs for a particular application, a plurality of different repeating units is preferably used thus obtaining copolymers, terpolymers and higher homologues. In order to substantially retain the service life enhancing property of the polymers comprising the repeating unit of the formula (C1), the proportion should be preferably at least 0.35.

In a particular embodiment, the polymer in accordance with the invention has, apart from the repeating unit of formula (C1) at least one further type of 1,4-phenylenevinylene repeating unit. Use of copolymers, terpolymers etc. of 1,4-phenylenevinylene repeating units in EL devices is attractive since they combine a high luminescence efficiency with good electron- and hole-transporting and injecting properties. Moreover they can be processed in a simple manner from solution.

If the polymer comprises a plurality of repeating units thus obtaining copolymers, terpolymers and the like it is most preferred that each of said units is a repeating unit of the formula (C1) or preferred embodiments thereof.

In accordance with WO 99/21936, in order to reduce red shift the further 1,4-phenylenevinylene repeat unit comprises a 2-aryl-1,4-phenylene unit wherein aryl is a phenyl, naphthyl, or biphenyl group which may or may not be substituted and/or in which, optionally, one or more of the non-substituted aromatic carbon atoms are replaced by nitrogen atoms. Preferred in this respect are polymers wherein the further repeating unit comprises a 2-phenyl-1,4-phenylene or 2,5-diphenyl-1,4-phenylene unit wherein each phenyl group is optionally substituted by one or more substituents R, wherein R has the same meaning as in formula (C1) and each occurrence of R involves a selection independent of the other occurrences.

With respect to suitable methods of preparing the polymers in accordance with the invention and monomers used to prepare said polymers reference is made to methods disclosed in the international applications WO 98/27136 and WO 99/21936 which are hereby incorporated by reference.

The invention further relates to the use of a polymer in accordance with the invention in an organic electroluminescent device.

In a preferred embodiment of said use, the device is operated at a brightness of at least 100 Cd/m² or even at least 200 Cd/m².

In a more preferred embodiment of said use, the device is operated at a brightness of at least 100 Cd/m² or even at least 200 Cd/m² for at least 45 h or better at least

200 h. This in particular applies to the use of a yellow to green light emitting polymer. In another preferred use, the polymer is a red to orange light emitting polymer used in a EL device operated for at least 800 to 1200 h at a brightness of at least 100 Cd/m².

A particular embodiment relates to the use wherein the organic
5 electroluminescent device is operated such that the temperature of the device is at least 5 to 10 °C above room temperature.

It has been found that if the brightness of organic EL device having a large light emitting surface area, say at least 10 to 20 cm², such as a back light for an LCD, and comprising a polymer of the type disclosed in WO 99/21936 is increased from 20 to 100 -
10 200 Cd/m², the temperature of the device increases by 5 to 10 °C. This is due to the heat generated by the device itself. Further experiments have shown that this seemingly small increase in temperature leads to a reduction in service life by a factor of as much as two. Since the polymers in accordance with the invention already have an excellent service life when driven at ambient temperatures of 70 to 80 °C, the polymers in accordance with the
15 invention can be suitably used in an EL device which is operated such that the device temperature is at least 5 to 10 °C above room temperature.

In a further aspect, the invention relates to an organic electroluminescent device comprising an aryl-substituted poly-p-arylenevinylene in accordance with the invention.

20 Preferably, the aryl-substituted poly-p-arylenevinylene in accordance with the invention is used as the electroluminescent material.

Apart from comprising the polymers in accordance with the invention, the EL device may be of a conventional type.

Generally, a conventional organic or, in particular polymer, EL device
25 comprises at least an organic or, in particular polymer, electroluminescent layer disposed between an electron-injecting electrode (~~an~~ a cathode) and a hole-injecting electrode (an anode). If a suitable voltage is applied to the electrodes, the organic EL layer emits light. Using different organic EL materials the color of the light emitted can be varied.

Preferably, the polymer in accordance with the invention is part of the material
30 from which the organic electroluminescent layer is made but it may also be part of other layers such as transport or injecting layers. Optionally, the organic EL material contains further substances, organic or inorganic in nature, which may be homogeneously distributed on a molecular scale or present in particulate form. In particular, compounds improving the charge-injecting and/or charge-transport capability of electrons and/or holes, compounds to

improve and/or modify the intensity or color of the light emitted, stabilizers, and the like may be present.

The organic EL layer preferably has an average thickness of 50 nm to 200 nm, in particular, 60 nm to 150 nm or, preferably, 70 nm to 100 nm.

5 The electron-injecting electrode is suitably made of a metal(alloy) having a low work function, such as Yb, Ca, Mg:Ag Li:Al, Ba or is a laminate of different layers such as Ba/Al or Ba/Ag electrode in which preferably the Ba layer is less than 10 nm thick.

 The hole-injecting electrode is suitably made of a metal (alloy) having a high work function such as Au, Pt, Ag. Preferably, a more transparent hole-injecting electrode
10 material, such as an indiumtin oxide (ITO), is used. Conductive polymers such as a polyaniline (PANI) and a poly-3,4-ethylenedioxythiophene (PEDOT) are also suitable transparent hole-injecting electrode materials. Preferably, a PANI layer has a thickness of 50 to 200 nm, and a PEDOT layer 100 to 300 nm.

 Generally the EL device is supported by a substrate. Suitable substrate
15 materials include quartz, ceramics, glass and synthetic resins which may or may not be flexible. Preferably, the substrate is transparent with respect to the light to be emitted. If a transparent hole-injecting electrode such as ITO is used the EL device is preferably supported by the substrate via this ITO hole-injecting electrode.

 Optionally, the EL device comprises additional layers disposed between the
20 electrodes. Such additional layers include hole-injecting and/or transport (HTL) layers and electron-injecting and/or transport (ETL) layers. In order to improve service life, luminous efficiency (e.g. expressed in Cd/A) and/or power efficiency (e.g. expressed in lm/W) EL devices which comprise a laminate of anode/HTL layer/EL layer/cathode, anode/EL layer/ETL layer/cathode, or anode/HTL layer/EL layer/ETL layer/cathode are preferred.

25 In addition to the polymers in accordance with the invention, suitable materials for the hole-injecting and/or hole-transport layers (HTL) include aromatic tertiary amines, in particular diamines or higher homologues, polyvinylcarbazole, quinacridone, porphyrins, phthalocyanines, poly-aniline and poly-3,4-ethylenedioxythiophene.

 In addition to the polymers in accordance with the invention, suitable
30 materials for the electron-injecting and/or electron-transport layers (ETL) are oxadiazole-based compounds and aluminiumquinoline compounds.

 If ITO is used as the anode, the EL device preferably comprises a 50 to 300 nm thick layer of the hole-injecting/-transport layer material poly-3,4-ethylenedioxythiophene or a 50 to 200 nm thick layer of polyaniline.

The EL device in accordance with the invention can be suitably used as a lighting or display device. Particularly, the EL device can be used as a back light for a liquid crystal display, as a segmented display device or a matrix display device of the passive or active type or a display in which several of these types of displays are combined. Both full-
5 color and monochrome display devices can be made.

A particular embodiment of the EL device in accordance with the invention is capable of providing a service life of at least 45 to 200 h when driven at a constant current, at an initial brightness of 200 Cd/m^2 , and at an ambient temperature of 80°C . In order to enable
10 to EL device to provide said capability it not only comprises a polymer in accordance with the invention as the EL material but is further specifically adapted by including a voltage source such as a battery capable of providing a voltage necessary to operate the device at a brightness of at least 200 Cd/m^2 .

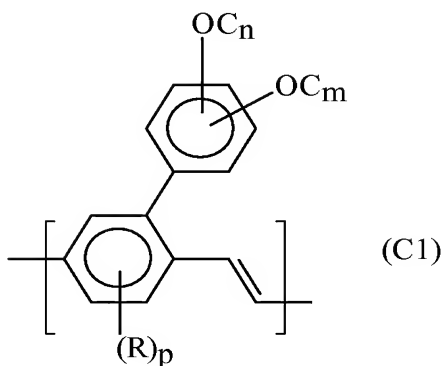
Another particular embodiment of the EL device in accordance with the invention comprises a red to orange light emitting aryl-substituted poly-p-arylenevinylene
15 and is capable of providing a service life of at least 800 to 1200 h when driven at a constant current, at an initial brightness of 100 Cd/m^2 , and at an ambient temperature of 70°C . In order to enable the EL device to provide said capability it not only comprises a polymer in accordance with the invention as the EL material but is further specifically adapted by including a voltage source capable of providing a voltage necessary to operate the device at a
20 brightness of at least 100 Cd/m^2 .

In another related aspect, the invention relates to an organic electroluminescent device comprising an organic electroluminescent, charge-transport and/or charge-injecting layer consisting of a material which, at least at one temperature in the range of 100 to 200°C , has a viscosity higher than or equal to the viscosity of an aryl-substituted
25 poly-p-arylenevinylene in accordance with the invention.

Surprisingly, it is found that the service life of an organic EL device depends on the viscosity of the material from which the organic layer is made. In particular, a high viscosity is found to increase the service life of the organic EL devices. Even more surprisingly it is found that this applies for organic EL devices driven at temperatures much
30 lower than the 100 to 200°C temperature range at which the viscosity is measured. Although in principle there is no reason to suppose that this dependency does not apply to all organic electroluminescent charge-transporting and/or charge-injecting materials used in organic EL devices it applies in particular if the material comprises a poly-p-arylenevinylene. More in particular it applies to the polymers in accordance with the invention.

-Since the temperatures experienced by EL devices are in practice generally lower than 100 °C, preferably, the viscosity near the lower end of the temperature range of 100 to 200 °C is higher than the viscosity of the aryl-substituted poly-p-arylenevinylenes in accordance with the invention.

Preferred embodiments of the EL device in accordance with the invention are ~~claimed in claims 11 and 12~~ An organic electroluminescent device comprising an organic electroluminescent, charge-transport and/or charge-injecting layer consisting of a material which, at least at one temperature in the range of 100 to 200 °C, has a viscosity higher than or equal to the viscosity of an aryl-substituted poly-p-arylenevinylene in which the Aryl-substituted poly-p-arylenevinylenes comprising a repeating unit of the formula (C1),



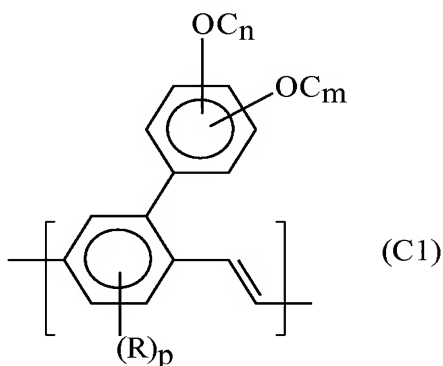
in which one or more of the unsubstituted aromatic carbon atoms may be replaced by nitrogen atoms, $-OC_m$ and $-OC_n$ are alkoxy groups, m and n are integers from 2 to 6 with $m + n = 8$, p is 0, 1, 2 or 3 and in which R is CN, Cl, F, CF_3 , NO_2 , or SO_3Z wherein Z is a monovalent cation such as Na^+ , or in which R is $-XR^1$ wherein the unit $-X-$ represents a single bond, $-O-$, $-S-$, $-CO-$, $-COO-$, $-OCO-$, $-SO-$, $-SO_2-$, $-N(R^2)-$ or $-N(R^2)CO-$, and wherein R^1 and R^2 are the same or different and constitute a straight-chain branched or cyclic C_1 - C_{20} alkyl group or together an C_1 - C_{20} alkylene group, in which C_1 - C_{20} alkyl or C_1 - C_{20} alkylene group one or more hydrogens are optionally substituted by F or a C_4 - C_{12} aryl group and/or one or more non-adjacent $-CH_2-$ units are optionally substituted by C_4 - C_{12} arylene, $-O-$, $-S-$, $-CO-$, $-COO-$, $-OCO-$, $-SO-$, $-SO_2-$, $-N(R^3)-$ or $-N(R^3)CO-$ where R^3 is C_1 - C_{20} alkyl, or in which R is a C_4 - C_{12} aryl group which may or may not be substituted;

Or it is the preceeding such Aryl-substituted poly-p-arylenevinylenes wherein $-OC_m$ and/or $-OC_n$ is 2-methylpropyloxy;

Or it is any of the preceeding Aryl-substituted poly-p-arylenevinylenes wherein $-OC_m$ and/or $-OC_n$ is 2-methylpropyloxy

Or it is any of the preceeding Aryl-substituted poly-p-arylenevinylenes wherein the repeating unit (C1) is a 2-(3',4'-bis(2methylpropyloxy)phenyl)-1,4-phenylene-vinylene repeating unit.

-Another preferred embodiment is an organic electroluminescent device comprising an organic electroluminescent, charge-transport and/or charge-injecting layer consisting of a material which, at least at one temperature in the range of 100 to 200 °C, has a viscosity higher than or equal to the viscosity of an aryl-substituted poly-p-arylenevinylene in which the Aryl-substituted poly-p-arylenevinylenes comprising a repeating unit of the formula (C1),



in which one or more of the unsubstituted aromatic carbon atoms may be replaced by nitrogen atoms, $-OC_m$ and $-OC_n$ are alkoxy groups, m and n are integers from 2 to 6 with $m + n = 8$, p is 0, 1, 2 or 3 and in which R is CN, Cl, F, CF_3 , NO_2 , or SO_2Z wherein Z is a monovalent cation such as Na^+ , or in which R is XR^+ wherein the unit X represents a single bond, O, S, CO, COO, OCO, SO, SO_2 , $N(R^2)$ or $N(R^2)CO$, and wherein R^+ and R^2 are the same or different and constitute a straight chain branched or cyclic C_1 - C_{20} alkyl group or together an C_1 - C_{20} alkylene group, in which C_1 - C_{20} alkyl or C_1 - C_{20} alkylene group one or more hydrogens are optionally substituted by F or a C_4 - C_{12} aryl group and/or one or more non adjacent CH_2 units are optionally substituted by C_4 - C_{12} arylene, O, S,

~~CO, COO, OCO, SO, SO₂, N(R²) or N(R²)CO where R² is C₄-C₂₀-alkyl, or in which R is a C₄-C₁₂-aryl group which may or may not be substituted;~~

~~Or it is the preceeding Aryl-substituted poly-p-arylenevinylenes wherein -OC_m and/or -OC_n is 2-methylpropyloxy;~~

~~Or it is any of the preceeding Aryl-substituted poly-p-arylenevinylenes wherein -OC_m and/or -OC_n is 2-methylpropyloxy~~

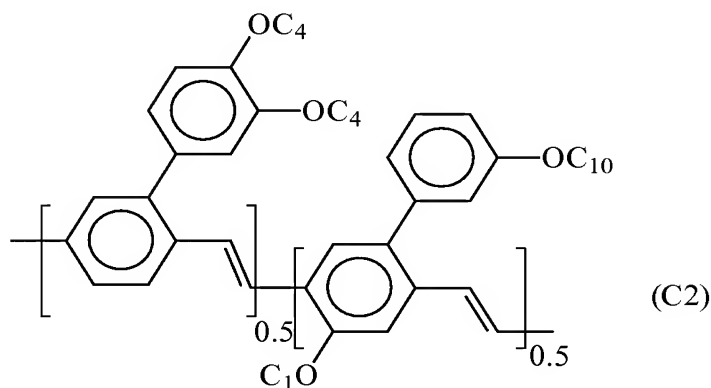
~~Or it is any of the preceeding Aryl-substituted poly-p-arylenevinylenes wherein the repeating unit (C1) is a 2-(3',4'-bis(2methylpropyloxy)phenyl)-1,4-phenylene-vinylene repeating unit. an organic electroluminescent, charge-transport and/or charge-injecting layer consisting of a material which, at least at one temperature in the range of 100 to 200 °C, has a viscosity higher than or equal to the viscosity of an aryl-substituted poly-p-arylenevinylene in which the aryl-substituted poly-p-arylenevinylenes comprising a repeating unit of the formula (C1), in which one or more of the unsubstituted aromatic carbon atoms may be replaced by nitrogen atoms, -OC_m and -OC_n are alkoxy groups, m and n are integers from 2 to 6 with m + n = 8, p is 0, 1, 2 or 3 and in which R is CN, Cl, F, CF₃, NO₂, or SO₂Z wherein Z is a monovalent cation such as Na⁺, or in which R is XR⁺ wherein the unit X represents a single bond, O, S, CO, COO, OCO, SO, SO₂, N(R²) or N(R²)CO, and wherein R⁺ and R² are the same or different and constitute a straight chain branched or cyclic C₄-C₂₀ alkyl group or together an C₄-C₂₀ alkylene group, in which C₄-C₂₀ alkyl or C₄-C₂₀ alkylene group one or more hydrogens are optionally substituted by F or a C₄-C₁₂-aryl group and/or one or more non-adjacent CH₂ units are optionally substituted by C₄-C₁₂-arylene, O, S, CO, COO, OCO, SO, SO₂, N(R²) or N(R²)CO where R² is C₄-C₂₀-alkyl, or in which R is a C₄-C₁₂-aryl group which may or may not be substituted. wherein m = n.~~

3. _____.

4. _____ Aryl-substituted poly-p-arylenevinylenes as claimed in claim 3 wherein the repeating unit (C1) is a 2-(3',4'-bis(2-methylpropyloxy)phenyl)-1,4-phenylene-vinylene repeating unit.

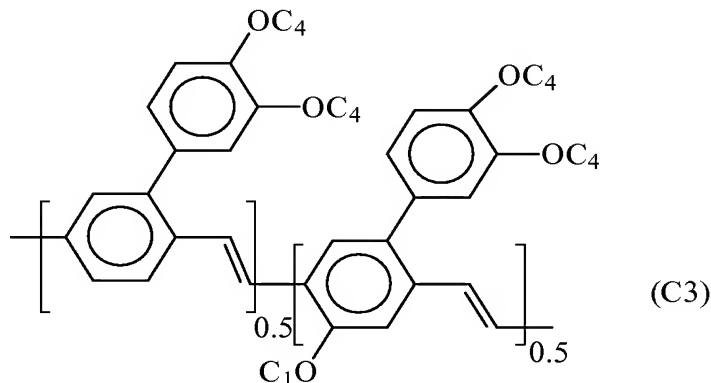
An organic electroluminescent device as claimed in claim 10 characterized in that the material from which the electroluminescent charge transport and/or charge injecting layer is made has, at least at one temperature in the range of 100 to 200 °C, a viscosity which is higher than or equal to the viscosity of the polymer of the repeating unit according to the formula (C2)

where -OC₁₀ is 3,7-dimethylhexyloxy dimethylhexyloxy octyloxy and -OC₄ is 2-



methylpropyloxy.

Yet another preferred embodiment of the invention is an An organic electroluminescent device as claimed in claim 11 characterized in that the material from which the electroluminescent, the charge-transport and/or the charge-injecting layer is made has, at least at one temperature in the range of 100 to 200 °C, a viscosity which is higher than or equal to the viscosity of the polymer of the repeating unit according to the formula (C3)



where -OC₄ is 2-methylpropyloxy.---

Many methods of measuring the viscosity are known in the art and may be used to measure the viscosity of the material.

In the context of the invention it is preferred to use a method which measures a viscosity-related parameter. Hereinafter this viscosity-related parameter is referred to as the viscosity η . This preferred method comprises the steps of:

- a) providing, on a substrate, a film of the material of thickness h_0 and providing a steel ball having a radius R ;
- b) bringing both the film and the ball to a temperature T within a range of 100 to 200 °C;
- c) positioning the ball on a surface of the film facing away from the substrate and pressing the ball into the film in a direction at right angles to the substrate with a force F during a time t , thus producing a circular indentation, if any, in the film;
- d) removing the steel ball and measuring the radius of indentation r of the circular indentation;
- e) determining the viscosity η from the equation (A1)

$$\frac{Ft}{6\pi\eta R^2} = \frac{3}{2} + \frac{1}{2(h/h_0)^2} - \frac{2}{(h/h_0)} - \log(h/h_0) \quad (A1)$$

wherein h is determined from the equation (A2),

$$r = \sqrt{4Rh_0(1 - (h/h_0))} \quad (A2), \text{ and}$$

h_0 is the initial film thickness,

- f) optionally, repeating steps a)-e) using a different temperature T .

The ball is to be removed slowly so as to avoid viscous fingering. The magnitude of the viscosity is in general such that the material flows back very slowly allowing enough time for an accurate measurement of the radius of indentation.

An important advantage of this method is that material of which the viscosity is to be determined can be provided in the form of a layer. The method can even be used for very thin layers ($< 1 \mu\text{m}$) which also demonstrates that only a very small amount of material is necessary to obtain a measurement. The value of the viscosity is accurate within a factor of 2.

The method being suitable to determine the viscosity of thin layers and the EL device in accordance with the invention comprising such a thin layer of organic material, the

EL device itself can be used as the sample for determining the viscosity which makes the method convenient for for example quality control purposes. In order to render the EL device suitable for measuring the viscosity, layers covering the organic layer or layers such as the cathode layer may have to be removed.

5

These and other aspects of the invention will be apparent from and elucidated with reference to the examples described hereinafter.

In the drawings:

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The sole figure schematically shows, in a cross-sectional view, an organic EL device.

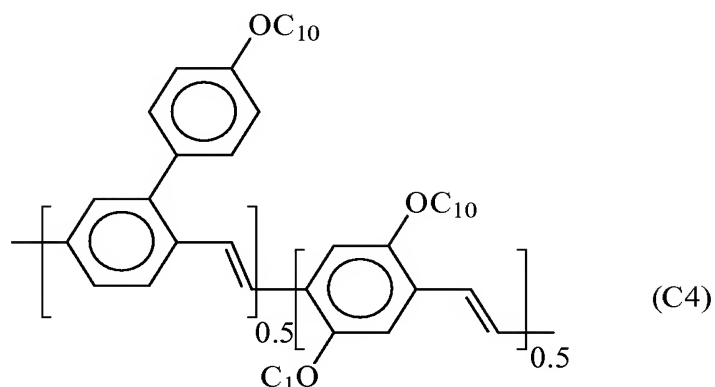
Comparative example 1

(red to orange light emission)

15

Referring to Fig. 1, in this comparative example, an electroluminescent device 1 comprises a substrate 3 of sodalime glass coated with a layer of an indiumtin oxide (ITO) supplied by Balzers which is in turn covered by a 250 nm layer of the hole-injecting material poly-3,4-ethylenedioxythiophene (supplier Bayer) provided by means of spin-coating. The latter two layers together constitute the hole-injecting electrode 5. The electroluminescent layer 7 is also provided by means of spin-coating and is made of a material consisting of an aryl-substituted poly-p-arylenevinylene of the repeating unit of formula (C4)

20



wherein OC₁₀ is 3,7-dimethyloctyloxy and which is a polymer not in accordance with the invention. The polymer C4 is disclosed in WO 99/21936 and photo- and electroluminesces red to orange light. The EL layer 7 is covered, in succession, with a Ba and Al layer which

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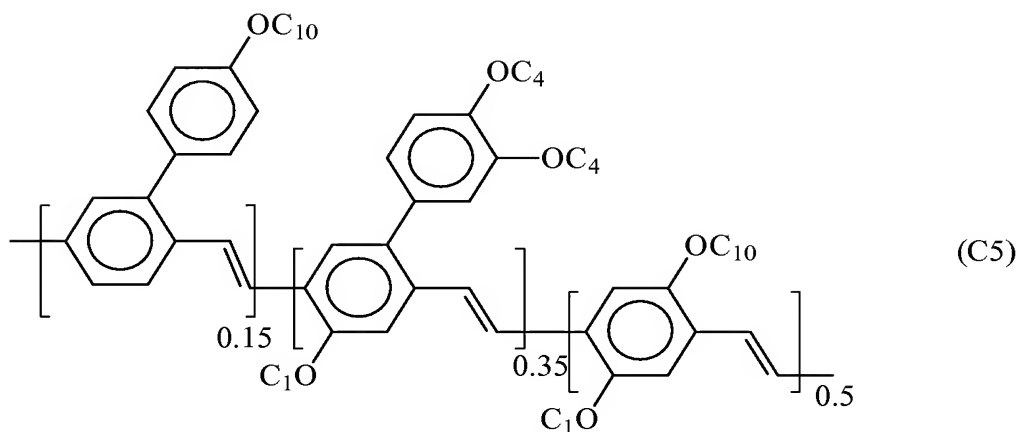
together ~~from~~ form the electron-injecting electrode 9 each layer being applied by means of deposition of metal vapor in vacuo. The EL device 1 further includes a voltage source 11 capable of providing a voltage sufficient to obtain an emission of light of brightness 100 Cd/m² or more. The EL device 1 is encapsulated so as to avoid contact with oxygen and water.

The EL device 1 is subjected to a service life test in which the device, the positive pole of the voltage source being connected to the anode 5 and the negative electrode to the cathode 9, is driven at constant current and at an initial brightness of 100 Cd/m² while maintaining an ambient temperature of 70 °C. The service life, defined as the time within which the brightness drops to half its initial value, thus determined is about 180 h. The voltage increase needed to maintain a constant current during the service life is about 0.015 V/h. By comparison, according to WO 99/21936, the service life at 20 Cd/m² and room temperature exceeds 5000 h which demonstrates the drastic reduction of service life associated with the use of a higher brightness and an elevated ambient temperature of 70 °C.

Example 1

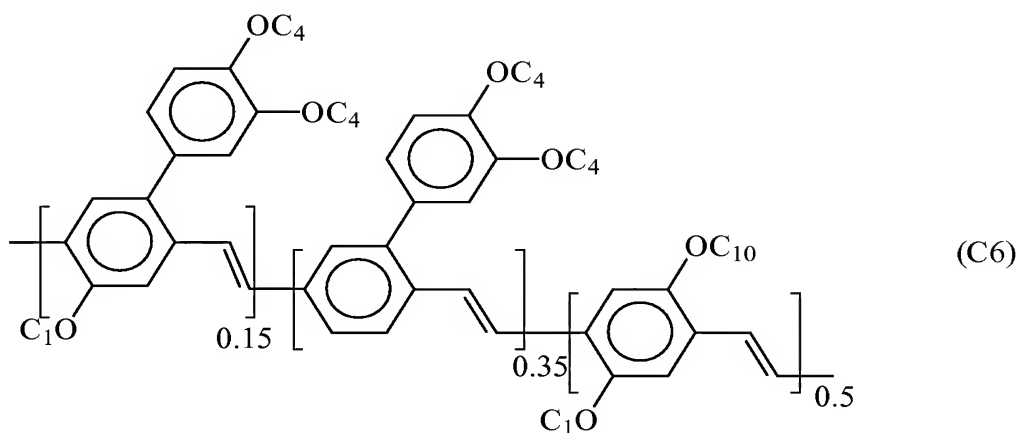
(red to orange light emission)

The previous example is repeated with the difference that the EL layer 9 is made of an aryl-substituted poly-p-arylenevinylene polymer in accordance with the invention, said polymer being a polymer of the repeating unit of the formula (C5) or (C6)



The polymers of formula (C5) and (C6) are synthesized according to processes analogous to processes disclosed in WO 98/27136 and/or WO 99/21936 which are hereby incorporated by reference. The monomers (intermediate compounds) corresponding to the different repeating units are disclosed in said international applications.

5 The EL devices based on the polymers of formula (C5) and (C6) emit red to orange light.



10 When subjected to the service life test of the comparative example, the service life of the EL device comprising the polymer of formula (C5) is 925 h. The voltage increase is about 2 mV/h. By collecting the data for a large number of samples the service life is found to be at least 800 to 1200 h.

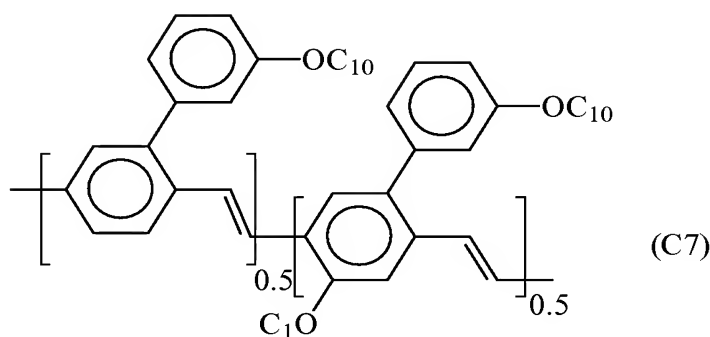
15 Similarly, the service life of the EL device comprising the polymer of formula (C6) is also at least 800 to 1200 h. The voltage increase is about 1.5 to 2 mV/h.

This example clearly demonstrates that the service life increases significantly by using an aryl-substituted poly-p-arylenevinylene in accordance with the invention. In particular, it demonstrates the use of a red to orange light emitting polymer in accordance with the invention in an organic EL device capable of providing a service life of at least 800 to 1200 h when driven at a constant current, an initial brightness of 100 Cd/m², and an ambient temperature of 70 °C.

Comparative example 2

(yellow to green light emissive polymers)

5 Comparative example 1 is repeated with the difference that the polymer is replaced by a polymer not in accordance with the ~~subject matter of the claims~~ invention, and emitting yellow to green light, this polymer being a polymer of repeating unit of formula



(C7)

wherein OC₁₀ is 3,7-dimethyloctyloxy.

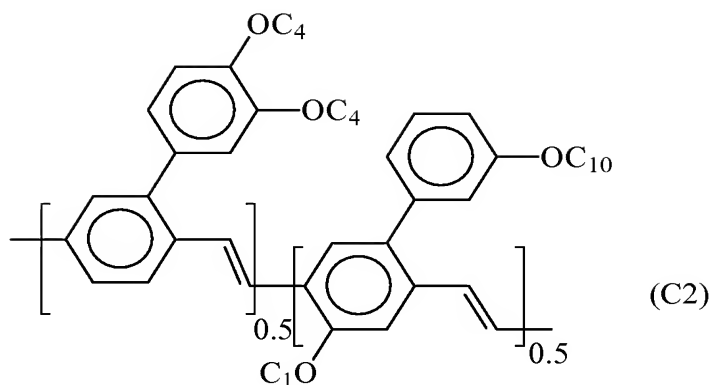
10 The device thus obtained is subjected to a service life test similar to that of comparative example 1, in which the EL device is driven at constant current and at an initial brightness of 200 Cd/m² while maintaining an ambient temperature of 80 °C. The service life, defined as the time within which the brightness drops to half its initial value, thus determined is about 10 h. The efficiency of the EL device is 9.0 Cd/A.

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Example 2

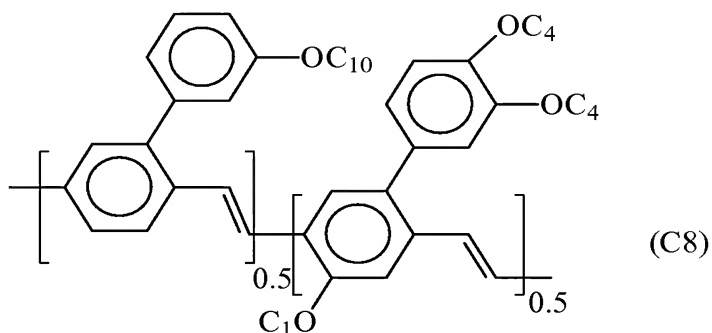
(yellow to green light emissive polymers)

20 Comparative example 2 is repeated with the difference that the polymer is replaced by a polymer in accordance with the ~~subject matter of the claims~~ invention, emitting yellow to green light, this polymer being a polymer of repeating unit of formula (C2)



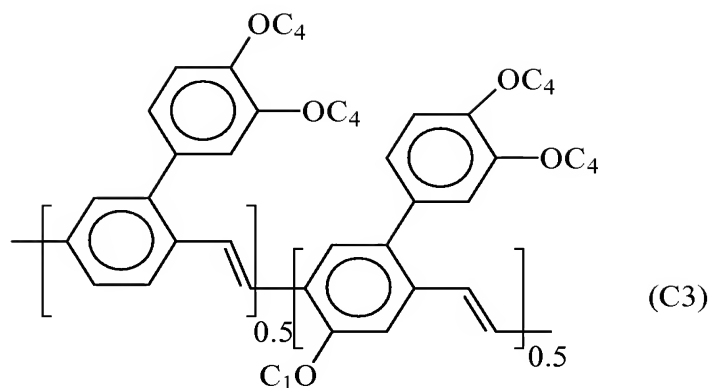
wherein OC₁₀ is 3,7-dimethyloctyloxy and OC₄ is 2-methylpropyloxy. When the EL device thus obtained is subjected to the service life test of comparative example 2, in which the EL device is driven at constant current and at an initial brightness of 200 Cd/m² while maintaining an ambient temperature of 80 °C, the service life, defined as the time within which the brightness drops to half its initial value, is about 55 h, whereas the efficiency of the EL device is 9.0 Cd/A.

Similarly, the EL device comprising the yellow light-emitting polymer of repeating unit according to formula (C8)



as the EL material has a service life of 70 h and an efficiency of 9.0 Cd/A.

The EL device comprising the yellow light-emitting polymer of repeating unit of formula (C3) as the EL material has, when subjected to the same accelerated service life test, a service life of 200 h and an efficiency of 9.0 Cd/A.



Example 3

The viscosity of the polymer with repeating unit according to the formula (C7) is measured as follows:

- An ITO-coated glass substrate is provided with a layer of the polymer by means of spin-coating. The thickness h_0 of the layer is 266 nm.
- The substrate thus obtained is mounted on a base plate provided with a heating foil and a thermocouple allowing the temperature of the substrate to be controlled. The temperature of the polymer layer is monitored using a thermocouple glued thereon.
- A steel ball having a radius R of 1.50 mm is polished to optical quality and fitted with a heating element in the form of NTC resistor and thermocouple.
- The steel ball and the substrate are then each brought to a temperature T of 100 °C and stabilized at that temperature for about 10 min.
- With the substrate resting on the base plate, the steel ball is carefully positioned on the surface of the polymer layer facing away from the substrate so as to avoid strong impact.
- For a loading time t of typically 60 to 600 s the ball is left positioned on the substrate. The ball exerts a gravitational force on the layer which, due to the viscous nature of the layer, makes it sink slowly into the layer thus producing a circular indentation. The force F which is exerted by the ball on the film is 0.63 N.
- After the loading time t has elapsed the steel ball is removed slowly so as to avoid viscous fingering.
- A new measurement is made by positioning the ball about a quarter mm away from the indentation. This process is repeated a number of times at different temperatures in the range of 100 to 200 °C.

- Of each of the circular indentations thus produced the radius of indentation r is measured using a microscope provided with an interference contrast facility, and the viscosity η is determined from the equation (A1)

$$\frac{Ft}{6\pi\eta R^2} = \frac{3}{2} + \frac{1}{2(h/h_0)^2} - \frac{2}{(h/h_0)} - \log(h/h_0) \quad (A1)$$

5 wherein h is determined from the equation (A2),

$$r = \sqrt{4Rh_0(1 - (h/h_0))} \quad (A2), \text{ and}$$

h_0 is the initial layer thickness.

10 The magnitude of the viscosity is in general such that the material flows back very slowly allowing enough time for an accurate measurement of the radius of indentation after all the indentations have been made.

At large loading times t and temperatures T it may happen that the ball sinks all the way through the film. In this case the film thickness h_0 can be determined. The thickness thus obtained corresponds well to the thickness determined using atomic force
15 microscopy (AFM).

Using the method described above which involves a film of 266 nm thickness, it is found that the relationship between $\log \eta$ and $1/T$ of the polymer with repeating unit according to formula (C7) is approximately linear. The viscosity at 100 °C is 1.10^7 Pa.s and at 160 °C about 1.10^5 Pa.s. At temperatures below 100 °C the ball does not produce a
20 measurable indentation.

The measurements are repeated using a 327 nm thick layer of the polymer with repeating unit according to the formula (C2). Again, an approximately linear relationship between $\log \eta$ and $1/T$ is observed. At 100 °C the viscosity is 5.10^7 Pa.s whereas at 160 °C it is about 5.10^5 Pa.s.

25 The measurements are repeated using the polymer with repeating unit according to the formula (C3). The viscosity of this polymer is so high that at no temperature below 200 °C the steel ball produces a measurable indentation.

The combined results of this example and example 2 demonstrate that, in accordance with the invention, in order to improve the service life of an EL device
30 comprising an organic layer, or in particular a polymer electroluminescent layer, the viscosity of the polymer must be high.